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# Finite-temperature properties of the Hubbard chain with bond-charge interaction

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We investigate the one-dimensional (1D) Hubbard model with an additional bond-charge interaction, recently considered in the description of compounds that exhibit strong 1D features above the temperature of ordered phases. The partition function of the model is exactly calculated for a value of the bond-charge coupling; the behavior of the specific heat and spin susceptibility as a function of temperature is derived at arbitrary filling, and particularly discussed across the occurring metal-insulator transition. The results show that the bond-charge terms weaken the spin excitations of the system.

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## I. INTRODUCTION

In recent years the discovery of materials that in some energy regimes exhibit a strong one-dimensional (1D) character has renewed the investigation of models of interacting electrons in low-dimensional lattices. Within this context, an increasing interest is nowadays devoted to the effects of unconventional correlation mechanisms, different from the usual charge-charge interaction terms between electrons on the same site ( $U$ ) and on neighboring sites ( $V$ ). In particular, models have been considered<sup>1-5</sup> which also account for the modification of the electron hopping motion by the presence of particles with opposite spins (correlated hopping); such kind of terms are also called *bond-charge* interactions, since they actually describe the interaction of charges located on bonds with those that are located on the lattice sites.

The first field of application of such kind of models in condensed matter was the description of  $\pi$  electrons in conducting polymers such as polyacetylene  $(\text{CH})_x$ ; in particular it has been found<sup>4</sup> that, according to the strength of bond-charge coupling the dimerization of the polymeric chain can be enhanced or destroyed.

More recently, bond-charge models have been considered<sup>5</sup> to explain the rich temperature-pressure phase diagram observed for the Bechgaard salts,<sup>6</sup> i.e., the linear chain organic compounds such as tetramethyletraselenafulvalene  $(\text{TMTSF})_2X$  and tetramethyltetrathiafulvalene  $(\text{TMTTF})_2X$ , where  $X = \text{ClO}_4$  or Br. Indeed, for these materials it has been noticed that the spin-density wave (SDW) and the superconducting (SC) phases are adjacent, the symmetry of the SC order parameter being of  $p$ -wave character, rather than  $d$  wave as in cuprates. Recent studies have suggested that the presence of bond-charge terms (whose coupling constants may depend on the pressure) could explain the interplay between the  $p$ -wave SC and the SDW orders, with varying pressure and electron density  $\rho$  (filling).

This idea has yielded a remarkable effort to the investigation of electron models with correlated hopping terms, through both analytical and numerical methods. In particular, results have been obtained for the ground state<sup>7,8</sup> as well as for the low-temperature limit, by the method of bosonization,<sup>1,5</sup> which has allowed to sketch out the phase diagram with respect to the bond-charge coupling constant.

Despite such remarkable results, a satisfactory comparison with experimental data on Bechgaard salts has not been

achieved yet. This is mainly due to the fact that, at very low temperatures, 2D and 3D couplings between organic chains become relevant (see, for instance, the phase diagram in Ref. 9); this explains, in pass, the occurrence of ordered phases in these compounds. As a consequence, a one-dimensional picture for these materials is reasonable only above some reference temperature (of the order of  $10^2$  K), which of course also depends on the pressure. In order to compare theoretical results with experimental observations on Bechgaard salts, it is therefore necessary to examine such models at higher temperature, or to test whether the low-temperature range tractable through bosonization has a nonvanishing overlap with the 1D region of the  $P$ - $T$  phase diagram of such materials.

The purpose of the present paper is to investigate the properties for a model of bond-charge interaction at finite and arbitrary temperature. In particular, for one value of the bond-charge coupling, we shall derive the exact behavior of thermodynamic observables such as the specific heat and the spin susceptibility, and discuss how the correlated hopping terms affect the obtained shape.

## II. MODEL: SPECTRUM AND PARTITION FUNCTION

The bond-charge model we shall discuss reads as follows:

$$\hat{\mathcal{H}} = -t \sum_{\langle i,j \rangle, \sigma} [1 - x(\hat{n}_{i\bar{\sigma}} + \hat{n}_{j\bar{\sigma}})] c_{i\sigma}^\dagger c_{j\sigma} + U \sum_{i=1}^L \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} - h \sum_{i=1}^L (\hat{n}_{i\uparrow} - \hat{n}_{i\downarrow}) + \mu \sum_{i=1}^L (\hat{n}_{i\uparrow} + \hat{n}_{i\downarrow}). \quad (1)$$

In Eq. (1)  $c_{i\sigma}^\dagger, c_{i\sigma}$  are fermionic creation and annihilation operators on a one-dimensional chain of (say)  $(\text{TMTTF})_2X$ . Let each site  $i$  represent an adequately chosen unit cell<sup>10</sup> and  $L$  be the total number of sites;  $\sigma = \uparrow, \downarrow$  is the spin label,  $\bar{\sigma}$  denotes its opposite,  $\hat{n}_{j\sigma} = c_{j\sigma}^\dagger c_{j\sigma}$  is the electron charge with spin  $\sigma$ , and  $\langle i, j \rangle$  stands for neighboring sites. At each site  $i$  four possible states are possible, which we shall denote as follows:  $|\uparrow\rangle_i = c_{i\uparrow}^\dagger |0\rangle$ ,  $|\downarrow\rangle_i = c_{i\downarrow}^\dagger |0\rangle$ ,  $|0\rangle_i = |0\rangle$ ,  $|\downarrow\uparrow\rangle_i = c_{i\downarrow}^\dagger c_{i\uparrow}^\dagger |0\rangle$ .

The term in the first line of Eq. (1) is the hopping term, and, in particular, the parameter  $x$  represents the bond-charge coupling constant (for  $x=0$  the ordinary Hubbard model<sup>11</sup> is recovered); the three terms in the second line, respectively, describe the usual on-site Coulomb repulsion, a possible cou-

pling to an external magnetic field, and the chemical potential. Notice that, similarly to Refs. 2, 7, and 8 we do not consider here the neighboring site charge-charge interaction  $V$ , since its presence can be accounted for—in a first approximation—through a renormalized value of  $U$ .

The model (1) is rather general, and is expected to capture the main effects of bond-charge terms. In the present work we shall provide exact results for the value,

$$x = 1. \quad (2)$$

For this value of the coupling constant and for zero magnetic field ( $h=0$ ), the exact ground phase diagram as a function of  $U$  and the filling  $\rho$  was obtained in Refs. 7,8 for open and periodic boundary conditions, respectively. This result was derived noticing that, for  $x=1$ , (a) the term in  $U$  commutes with the hopping term; (b) the hopping term naturally allows a separation of the four possible states defined above into two groups, namely,  $A=\{|\uparrow\rangle, |\downarrow\rangle\}$  and  $B=\{|0\rangle, |\downarrow\uparrow\rangle\}$ . In fact, for such value of the bond-charge coupling, the hopping term actually *permutes*  $A$  states with  $B$  states only, but not  $A$  (or  $B$ ) states between themselves.

In Ref. 12 these arguments have been generalized. In the first instance, further commuting terms, other than the on-site Coulomb repulsion, such as the magnetic field, can be added to the hopping part. Secondly, it has been pointed out that the properties of the hopping term of Eq. (1) are shared by a whole subclass of extended Hubbard models, to which the Hamiltonian (1) belongs. More explicitly, each model within this subclass identifies a specific number and set of Sutherland species, i.e., the *groups* of states such that the Hamiltonian only permutes the states related to different species, leaving unaltered neighboring states that belong to the same species; for this reason the models of this subclass have been termed “generalized permutators.”<sup>12</sup> The number  $n$  of Sutherland species is by definition not greater than the number of physical states (4 in the case of a single orbital). For model (1) the Sutherland species are 2, and precisely  $A$  and  $B$ .

Recognizing that a model identifies (up to some commuting terms) a set of Sutherland species greatly simplifies the calculation of the partition function. The crucial point that allows the use of *open* boundary conditions, instead of the customary periodic ones; although in the thermodynamic limit the bulk properties are not affected by either choice, the calculations are more straightforward for the former. Indeed in an open one-dimensional chain the set of eigenvalues of a generalized permutator is equal to that of an ordinary permutator between  $n$  objects, i.e., the effective dimensionality of the Hilbert space is reduced (reduction theorem). As a consequence of that, the degeneracy of the eigenvalues can also be computed, simply counting the ways one can realize a given configuration of Sutherland species. Such observations are rather general and have been used, for instance, to derive the exact thermodynamics of an extended Hubbard model of the above subclass.<sup>13</sup> We shall apply them here to obtain the partition function of the bond-charge model (1) for the value (2).

The reduction theorem is proved when realizing that, according to what observed above, the relative order of any sequence of states belonging to the same species is

preserved;<sup>7</sup> the Hamiltonian can therefore be diagonalized within each subspace of given set of sequences. In the case of model (1), the sequences  $\mathcal{S}_A$  and  $\mathcal{S}_B$  of  $A$  species and  $B$  species are separately preserved. Moreover, since in this case one has *two* Sutherland species, each invariant subspace is in a one-to-one correspondence with the states of a spinless fermion (SF) space; if the  $A$  species relates to occupied and the  $B$  species to empty sites of the SF space, the form of an effective Hamiltonian for the SF problem is that of a tight-binding model, for the latter can be regarded to as a permutation between occupied and empty sites. The eigenvalues therefore read  $-2t \sum_{i=1}^L \cos k n_k^A$ , with  $n_k^A$  quantum numbers valued 0 or 1, and  $k = \pi n / (L+1)$ , with  $n = 1, \dots, L$ . The number of SF equals that of  $A$ -species objects ( $N_A$ ) and one has  $\sum_k n_k^A = N_A$ ; the number of empty sites is then  $N_B = L - N_A$ . Under open boundary conditions, the specific sequence is irrelevant to the action of the hopping term, so that all the subspaces share the same spectrum; in general this does not hold under periodic boundary conditions (see, for instance, Refs. 8 or 14). The Fock space is thus reorganized in terms of states defined by species  $A$  and  $B$  and their related degeneracy.

The inclusion of the further (commuting) terms simply lifts the degeneracy of the eigenvalues of the hopping term, yielding a spectrum that depends, apart from  $\{n_k^A\}$ , also on  $N_{\downarrow\uparrow}$  and on  $N_{\uparrow}$ , the latter being the eigenvalues of the operators  $\sum_{i=1}^L \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}$  and  $\sum_{i=1}^L \hat{n}_{i\uparrow} (1 - \hat{n}_{i\downarrow})$ . The spectrum of model (1) thus reads

$$E = E(\{n_k^A\}; N_{\downarrow\uparrow}; N_{\uparrow}) \\ = \sum_k (\epsilon_k - \mu + h) n_k^A + (U - 2\mu) N_{\downarrow\uparrow} - 2h N_{\uparrow}, \quad (3)$$

where  $\epsilon_k = -2t \cos k$ ; the identities  $N_{\uparrow} - N_{\downarrow} = 2N_{\uparrow} - N_A$  and  $N = N_{\uparrow} + N_{\downarrow} = N_A - 2N_{\downarrow\uparrow}$  have been exploited. In the case of zero magnetic field ( $h=0$ ) one recovers the spectrum that was minimized in Ref. 7 at fixed number of particles ( $\mu = \text{const} = 0$ ) to obtain the ground-state phase diagram  $U$  vs  $\rho$ .

The degeneracy  $g$  corresponds to the different ways one can realize a configuration of Sutherland species, with the constraint that the total numbers  $N_{\downarrow\uparrow}$  and  $N_{\uparrow}$  appearing in Eq. (3) remain unchanged; a simple calculation yields

$$g(E(\{n_k^A\}; N_{\downarrow\uparrow}; N_{\uparrow})) = \binom{L - N_A}{N_{\downarrow\uparrow}} \binom{N_A}{N_{\uparrow}}. \quad (4)$$

The rearrangement of the Fock space deriving from the identification of the Sutherland Species allows a straightforward calculation of the (gran-canonical) partition function

$$\mathcal{Z} = \sum_{\{n_k^A\}} \sum_{N_{\downarrow\uparrow}=0}^{L-N_A} \sum_{N_{\uparrow}=0}^{N_A} g(E) e^{-\beta E(\{n_k^A\}; N_{\downarrow\uparrow}; N_{\uparrow})} \\ = (1 + e^{\beta(\mu - U/2)})^L \prod_{k=1}^L (1 + e^{-\beta\{\epsilon_k - \mu^*(\mu, \beta, U, h)\}}). \quad (5)$$

In the second line of Eq. (5) we have defined

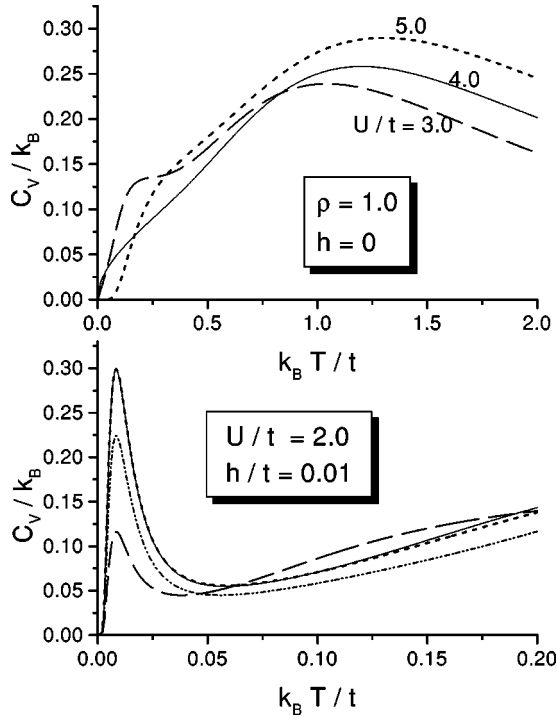


FIG. 1. The specific heat as a function of temperature. Top:  $\rho = 1$ ,  $h = 0$ : the metal-insulator transition is revealed through the change in low-temperature behavior from linear to exponential across the critical value  $U = 4t$ . Bottom:  $C_V$  for different filling values:  $\rho = 0.25$  (dashed),  $\rho = 0.50$  (dot-dashed);  $\rho = 0.75$  (dotted) and  $\rho = 1$  (solid); a low-temperature sharp peak emerges for nonvanishing magnetic field.

$$\mu^*(\mu, \beta, U, h) = \mu + \frac{1}{\beta} \ln \frac{2 \cosh \beta h}{1 + \exp 2\beta(\mu - U/2)}, \quad (6)$$

$\beta = 1/(k_B T)$  being the inverse temperature. Notice also that the product over  $k$  resulting in Eq. (5) is in form similar to the partition function of a tight-binding model of spinless fermions, where  $\mu^*$  plays the role of an effective chemical potential renormalized by the interaction  $U$ , the magnetic field  $h$  and the temperature itself.

### III. RESULTS AND DISCUSSION

By means of the partition function (5) derived in the preceding section, one can calculate the thermodynamic observables from the grand potential (per site)  $\omega = -\lim_{L \rightarrow \infty} k_B T \ln \mathcal{Z}$ . In doing that, we have eliminated the chemical potential  $\mu$  in favor of the filling through the relation  $\rho = \partial \omega / \partial \mu$ , as usual.

In Fig. 1 we have plotted the specific heat (per site)  $C_V$  as a function of the temperature. In particular, in the top figure we have examined the case of half-filling (i.e.,  $\rho = 1$ ) and zero magnetic field ( $h = 0$ ), for different values of the on-site Coulomb repulsion  $U$ . One can observe that, across the value  $U/t = 4$ , the low-temperature behavior of  $C_V$  changes from linear to exponential; explicitly, for  $U < 4t$  we have

$$C_V \sim \gamma T \quad \text{with} \quad \gamma = \frac{k_B^2 \pi}{6t \sqrt{[1 - (U/4t)^2]}}, \quad (7)$$

whereas for  $U > 4t$

$$C_V \sim k_B \frac{(U - 4t)^2}{8 \sqrt{\pi t^2}} (k_B T/t)^{-3/2} e^{-(U-4t)/2k_B T}. \quad (8)$$

This is a finite-temperature effect of a metal-insulator transition, in accordance with the result obtained in Refs. 7 and 8, where a charge gap  $\Delta_c = U - 4t$  is shown to open in the ground state for  $U > 4t$ . We recall that for  $x = 0$  (i.e., for the ordinary 1D Hubbard model) no metal-insulator transition occurs; the bond-charge term thus seems to give rise to a finite critical value  $U_c$ , increasing from 0 to  $4t$  as the coupling  $x$  is varied from 0 to 1. It is also worth emphasizing that such effect is opposite to the case of higher dimension, where the bond-charge interaction is found<sup>2</sup> to lower the critical value of the metal-insulator transition (Gutzwiller approximation, exact in the limit  $D \rightarrow \infty$ ).

Notice that, the ratio  $U/t$  is expected to scale inversely with the pressure, since the increase of the latter roughly enhances the hopping amplitude; as a consequence, the passage from a metallic to an insulating state with increasing  $U/t$  is in accordance with the qualitative features of the Bechgaard salts phase diagram.<sup>9</sup>

In the bottom Fig. 1,  $C_V$  is plotted for different filling values, fixed ratio  $U/t = 2$  and magnetic field  $h/t = 0.01$ . A sharp low-temperature peak, located at  $k_B T \sim h$ , is observed to emerge as soon as the magnetic field is turned on. Interestingly, the peak becomes basically filling independent as  $\rho$  enters the range  $[\bar{\rho}, 2 - \bar{\rho}]$ , with  $\bar{\rho} = \cos^{-1}(-U/4t)/\pi$ . This amounts to the fact that, within this range of  $\rho$ , particles can be added to the system only in form of singlet pairs, in accordance with the features of the phase diagram in Ref. 7.

In addition, one can show that

$$\lim_{T \rightarrow 0} \lim_{h \rightarrow 0} C_V/T \neq \lim_{h \rightarrow 0} \lim_{T \rightarrow 0} C_V/T \quad (9)$$

differently from the ordinary Hubbard model, where the two limits are interchangeable.<sup>15</sup> At half-filling and for  $|U + |2h|| < 4t$ , for instance, one has  $C_V \sim \gamma T$  with

$$\gamma = \frac{k_B^2 (3 \ln^2 2 + \pi^2)}{6 \pi t \sqrt{\{1 - [(U + 2|h|)/4t]^2\}}}. \quad (10)$$

Comparing Eq. (10) to Eq. (7), one can realize that Eq. (9) holds. Similarly, the exponential behavior, occurring when the gap is open, is different; namely, for  $|U + |2h|| > 4t$

$$C_V \sim k_B \frac{(U + 2|h| - 4t)^2}{4 \sqrt{\pi t^2}} (k_B T/t)^{-3/2} e^{-(U+2|h|-4t)/2k_B T} \quad (11)$$

to be compared to Eq. (8).

In Fig. 2 the specific heat of model (1) for  $x = 1$  is plotted aside the case  $x = 0$  (i.e., the Hubbard model) for strong coupling, namely,  $U = 8t$ . Notice that the ordinary Hubbard model has a low-temperature peak, whose origin (see, e.g.,

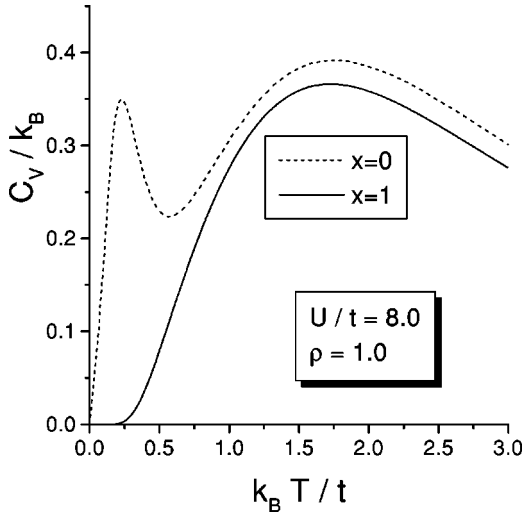


FIG. 2. The specific heat as a function of temperature for model (1) in the strong-coupling regime ( $U=8t$ ), at half-filling and zero magnetic field. The dotted line is the case  $x=0$ —i.e., the ordinary Hubbard model—obtained from Ref. 16, and the solid line the case  $x=1$ , obtained from our exact calculations. Continuity arguments suggest that the specific heat for arbitrary  $0 \leq x \leq 1$  lies between these two curves. The low-temperature peak originating from spin excitations is depleted by the bond-charge interaction.

Ref. 16) is due to spin degrees of freedom; the latter being not gapped, the low-temperature behavior of  $C_V$  is linear in spite of the fact that a charge gap is present at any  $U > 0$ .<sup>17</sup> In contrast, in model (1), for  $x=1$  the spectrum does not carry any spin quantum number, due to the rich symmetry of the model;<sup>18</sup> spins act therefore as a sort of dummy variables. Although the value  $x=1$  is a particular one, it is reasonable to expect that, for continuity argument, the plot of  $C_V$  for intermediate values  $0 \leq x \leq 1$  lies between the two curves. As a consequence, we can infer that the effect of spin excitations is weakened by the presence of the bond-charge interaction, at least in the strong-coupling regime.

In order to have a qualitative idea concerning the Bechgaard salts,<sup>9</sup> the temperature range of the figures compatible with the 1D regime of, e.g.  $(\text{TMTTF})_2\text{Br}$  is  $k_B T / t \geq 0.3$  (indeed  $t \sim 0.1$  eV and  $U \sim 1$  eV).

The depletion of spin excitation is also confirmed by the behavior of the magnetic susceptibility, defined as  $\chi = \mu_B^2 \langle \partial m / \partial h \rangle |_{h \rightarrow 0}$ . The calculation shows that  $\chi$  coincides with  $\mu_B^2 \rho_A / k_B T$ , where  $\rho_A$  (the density of A species along the chain) is a regular function of  $T$ , plotted at half-filling in Fig. 3. One can observe that, differently<sup>19</sup> from the ordinary Hubbard model ( $x=0$ ), the susceptibility is divergent for  $T \rightarrow 0$  with a Curie-law behavior, reminiscent of a system of independent magnetic moments. This also holds for any filling value, suggesting that, as  $x \rightarrow 1$ , the velocity of spin excitations vanishes, independently of  $U$  and of the filling; in particular, no spin gap is present for  $x=1$  either.

One can now compare such results with those obtained through a low-energy approach. The latter is reliable only when the interaction couplings and the thermal fluctuations

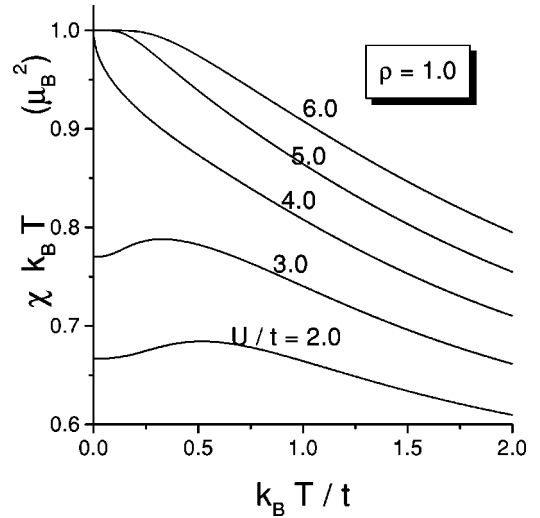


FIG. 3. The spin susceptibility of model (1) with  $x=1$  diverges like  $T^{-1}$  as  $T \rightarrow 0$ . The quantity  $\chi k_B T$  is the integral of the spin-spin correlation function, from fluctuation-dissipation theorem. A change in its low-temperature behavior is observed across the metal-insulator transition value.

are small compared with the bandwidth  $w=4t$ ; however, for the ordinary Hubbard model, one can (*a posteriori*) extrapolate the results concerning the formation of charge/spin gaps to the strong-coupling regime too.

Applying the bosonization technique to model (1), one can show (see also Refs. 1 and 5) that (a) the charge sector behaves like that of the Hubbard model, so that  $U_c=0$  at  $\rho=1$ , and (b) the spin sector is gapless for  $u'=U/t + 8x \cos(\pi\rho/2) > 0$ , the spin excitations exhibiting a velocity  $v_s = v_F \sqrt{1 - u'/\pi v_F}$ . In particular, a linear contribution to  $C_V$  from spin excitations is thus expected for any  $x$  as long as  $u' > 0$ .

The exact results of the present work (Figs. 2 and 3, and the discussion above) indicate that any attempt to mimic the ordinary Hubbard model extrapolating the low-energy approach to  $x \rightarrow 1$  would fail with respect to the spin channel, since the behavior is actually quite different at  $x \approx 1$ . This also holds for the charge channel, according to the results in Refs. 7 and 8 which show that for  $x=1$  the latter is gapless at half-filling for  $|U| \leq 4t$ .

In conclusion, in the present paper the finite-temperature properties of the Hubbard model with bond-charge interaction have been exactly derived for the value (2) of the bond-charge coupling. The results concerning the behavior of the specific heat and the magnetic susceptibility indicate that the bond-charge interaction tends to suppress the spin excitations of the system.

Previous investigations about such model were concerned with either the ground state or the low-temperature limit. In contrast, our calculations are valid at any temperature. We emphasize that for correlated quantum systems exact results are very rare at finite temperature, even for those models that have been proved to be integrable.



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